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Theoretical and calculational aspects of the radio refractive index of water vapor

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The radio refractive index is calculated by summing the refraction due to the infrared resonances of the water vapor monomer. Although this calculated refraction agrees well with measured values, its temperature dependence departs significantly from the measured variation with temperature. The rotational resonances of water vapor give a contribution to the radio refractive index that does not vary precisely as e/T^2 ; theoretical reasons are given for this departure from the classical refraction by permanent dipoles.

INTRODUCTION

The radio refractive index equation of moist air is commonly given [e.g., Thayer, 1974] by

$$(n - 1) \times 10^6 = K_1(P_a/T)Z_a^{-1} + K_2(e/T)Z_w^{-1} + K_3(e/T^2)Z_w^{-1} \quad (1)$$

where n is refractive index, P_a is partial pressure of dry air, e is partial pressure of water vapor, T is absolute temperature, and Z_a and Z_w are small correction factors for nonideal gas behavior. The expressions $(P_a/T)Z_a^{-1}$ and $(e/T)Z_w^{-1}$ are proportional to the number densities of dry-air molecules and water vapor molecules, respectively. The compressibility factors Z_a and Z_w are given by Owens [1967]. The coefficients K_1 , K_2 , and K_3 are obtained by fitting data to this equation. This traditional fit to terms of the form e/T and e/T^2 corresponds to the classical refraction by induced dipoles and permanent dipoles, respectively. We shall be concerned with the refraction caused by water vapor and hence with the corresponding coefficients K_2 and K_3 .

Each absorption resonance provides a nondispersive refraction at frequencies much lower than the resonance frequency. The electronic transitions at ultraviolet frequencies are the induced dipole type and hence provide refraction at optical frequencies that varies as P_a/T and e/T . Although pure vibrational transitions produce a radio frequency refraction that varies as (e/T) [Van Vleck, 1927], the infrared vibrational-rotational transitions do not entirely fit either classical analogy, and Zhevakin and Naumov [1967a] show that these resonances contribute little to the refractive index at radio frequencies. The far-infrared rotational transitions of the water molecule are the permanent-dipole type and can be expected to provide a radio refraction that principally varies as e/T^2 . However, Van Vleck [1932] shows that for the e/T^2 dependence to be valid it is vital that no permanent-dipole rotational transitions involve energy changes of the order of the thermal energy $K_B T$, where K_B is the Boltzmann constant and T is the absolute temperature. Equivalently, there must be no transitions having resonance frequency ν_i (in units cm^{-1}) of the order of $K_B T/hc$, where h is Planck's constant and c is the speed of light in vacuum. Note that $C \equiv K_B/hc = 0.695 \text{ cm}^{-1} \text{ K}^{-1}$. One cm^{-1} is 30 GHz. For typical atmospheric temperatures between 250 K and 300 K, the frequency CT lies between 175 and 210 cm^{-1} (5.25 THz and 6.3 THz). The rotational transitions of the water molecule certainly include strong resonances in and near this frequency interval. Thus there is theoretical expectation that the refractive index of water vapor may not be simply the sum of two terms behaving as e/T and e/T^2 .

Thayer [1974] hypothesized that the radio refractive index equation would be more accurate if K_2 were taken to have the same value as it does at optical frequencies. It is now clear that this is tantamount to assuming that the sum of refraction contributions from all the infrared water vapor resonances produces K_3 but does not change K_2 . We test this

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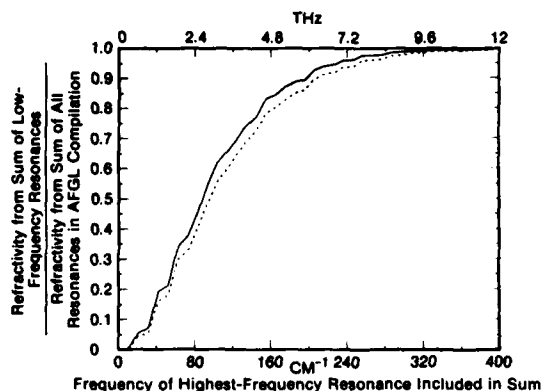


Fig. 1. The ratio of the radio refractivity contributed by all resonances having resonance frequency less than a value on the ordinate to the radio refractivity contributed by all infrared water vapor resonances. Solid curve corresponds to $T = 273$ K while the dashed curve is for $T = 333$ K.

hypothesis by calculating the sum of the contributions of infrared water vapor resonances to the refraction at radio frequencies.

CALCULATED REFRACTIVE INDEX

In principle, the refractive index can be calculated by summing the refraction contributions of all absorption resonances [Zhevakin and Naumov, 1967a, b; Birnbaum, 1953]. Birnbaum [1953] shows that for any sharp absorption resonance the Kramers-Kronig relationship implies a refraction in the far wings having a frequency dependence given by $n_i \propto 1/(v_i^2 - v^2)$, where n_i is the refractive index caused by the resonance labeled by the index i , v is frequency, and v_i is the resonance frequency. This result applies for either $v \ll v_i$ or $v \gg v_i$. Since this result applies to sharp absorption resonances in general, it certainly applies for any particular line shape model having a sharp absorption line shape (e.g., the Gross or Van Vleck-Weisskopf line shapes). Each resonance contributes a refraction n_i that is nondispersive in the limit of low electromagnetic frequency $v \rightarrow 0$; each line contributes an amount [Hill and Clifford, 1981]

$$n_i = S_i Q / 2\pi^2 v_i^2 \quad (2)$$

where S_i is the integrated absorption strength of the resonance, v_i is its line frequency, and Q is the number density of absorbing molecules. Any particular line shape model that is sufficiently sharp in the sense of Birnbaum [1953] (e.g., the Gross or Van

Vleck-Weisskopf line shapes) must give equation (2). The strongest (largest S_i) and lowest frequency (smallest v_i) resonances contribute most. Therefore, although the strong water vapor resonances in the vicinity of the frequency CT may not give a temperature dependence behaving like e/T^2 , the equally strong but lower frequency resonances may dominate the radio wave refraction and approximately restore this temperature dependence.

The line strength has temperature dependence arising from the partition function and the difference of two Boltzmann distributions. For the water molecule this temperature dependence is

$$S_i \propto T^{-3/2} [\exp(-E_i^L/CT) - \exp(-E_i^U/CT)] \quad (3)$$

where E_i^L and E_i^U are the values (in units cm^{-1}) of the lower and upper state energies responsible for resonance i and $C = 0.695 \text{ cm}^{-1} \text{ K}^{-1}$.

We have calculated the radio frequency refraction of the infrared resonances of water vapor by summing the contributions of all water vapor lines in the Air Force Geophysics Laboratory (AFGL) line parameter compilation. This compilation extends from lines at radio frequencies to those in the visible. All water vapor lines are included in the sum; no lines have been excluded, no matter how weak or how high in line frequency. The calculation was performed for several humidities from 0% to 100% for each of many temperatures from -10°C to 63°C and 110°C to 250°C and for total pressures of 0.2 atm, 0.8 atm, and 1.0 atm. As expected, for fixed temperature and humidity the refraction by water vapor was found to be independent of total pressure. Also as expected, the calculated refraction was proportional to the humidity. The Gross line shape having humidity and pressure-dependent line width was used, as by Hill *et al.* [1980]. This line shape gives (2) in the low-frequency limit. A 1-m radio wavelength was assumed; for such a low frequency there is no practical difference between the complete Gross formula and (2).

Suppose we begin summing the refraction with the lowest frequency in the line parameter compilation and proceed to include higher and higher frequency resonances. How does the refraction accrue as more resonances are included, and how high in frequency must we sum before the total refraction is approached? Figure 1 illustrates the answer by showing the ratio of the partial sum to the total sum. Although the graph only shows frequencies less than 400 cm^{-1} (i.e., 12 THz), the total sum includes all

lines from radio to visible frequencies. In principle, the curves in Figure 1 should step upward at the resonance frequency of each line; in fact, the curves are smoothed by plotting point to point at intervals of 10 cm^{-1} . Consider the proof by *Van Vleck* [1932] that rotational resonances should sum to provide a radio refraction that varies as (e/T^2) ; this proof requires neglecting $0.5(v_i/CT)^2$ relative to unity. If $0.5(v_i/CT)^2$ is less than 0.1, then $v_i < 85\text{ cm}^{-1}$ for $T = 273\text{ K}$. Figure 1 shows that less than half of the radio refraction arises from rotational resonances satisfying $0.5(v_i/CT)^2 < 0.1$. Thus we should not be surprised if the radio refraction caused by rotational resonances does not vary precisely as (e/T^2) .

In a sense, a measurement of radio refractive index is an empirical sum of the refraction contributions of all the infrared and ultraviolet resonances. It is natural to treat our calculated refractive indexes as a simulated set of experimental data. Thus we do as the experimentalists have done; that is, we fit the calculated refraction to the form $K_2(e/T)Z_w^{-1} + K_3(e/T^2)Z_w^{-1}$ as suggested by (1). This functional form fits the refraction very well; the rms error was only a few thousandths of one percent. The temperature range $0^\circ\text{--}60^\circ\text{C}$ corresponds to that used in the experiments by *Boudouris* [1963]; fitting to this temperature range gave the following infrared contributions to K_2 and K_3 :

$$K_2 = 33.30 \pm 1\text{ K/mbar} \quad (4)$$

$$K_3 = (3.5833 \pm 0.0032) \times 10^5\text{ K}^2/\text{mbar} \quad (5)$$

Equations (4) and (5) result from the sum of all infrared resonances in the AFGL compilation. Note that the total value of K_2 at radio frequencies is the sum of the above infrared contribution, 33.30 K/mbar , and the extrapolation to infinite wavelength of the empirically known value of K_2 at optical frequencies, $64.79 \pm 0.08\text{ K/mbar}$ [Thayer, 1974]; the latter is caused by the ultraviolet electronic resonances of the water molecule.

We examined fitting the calculated refraction to several other temperature ranges. For the temperature range $110^\circ\text{--}250^\circ\text{C}$, which roughly corresponds to the higher temperature range of experiments reviewed by *Birnbaum and Chatterjee* [1952], we found that the infrared contribution to K_2 is 2% larger than (4) and K_3 is 0.1% smaller than (5). For the temperature range -10° to 40°C , the infrared contribution to K_2 is 2% larger than (4), and K_3 is 0.05% smaller than (5). These deviations are within the error given in (4) and (5).

The choice of the estimated errors in (4) and (5) deserves explanation. Unlike experiments which have random variation, we are fitting to a smooth function of temperature. We must choose a nonstatistical measure of the error in K_2 and K_3 . Our choice is as follows: We first find the best fit that the computer can generate; then we make a small change in K_2 and refit by changing only K_3 , but not K_2 . We continue to change K_2 and refit K_3 until the rms error in the refraction is twice the rms error of the best fit. The errors stated in (4) and (5) are the difference between the values of K_2 and K_3 from the best fit and those that double the rms error in the refraction. This definition of error gives a conservative estimate that accounts for the fact that a decrease in K_2 can be approximately compensated by an increase in K_3 and vice versa. If all the rms error in the fit to the refraction were blamed on only one of K_2 or K_3 , but not the other, then the errors would be only about one-tenth the errors stated in (4) and (5). The difference between minimizing rms error as opposed to minimizing percent rms error changes K_2 and K_3 by about one-tenth the error stated in (4) and (5). The error in the fitting is not caused by computer round-off; it is caused by the computed refraction having temperature dependence that is not adequately described by (1).

The line strengths are given to three decimal places in the line parameter compilation. By allowing increases in K_3 to partially compensate for decreases in K_2 (and vice versa), three significant figures in the calculated refraction imply errors in K_2 and K_3 that are about 10 times larger than those stated in (4) and (5). As will be seen in the discussion section, such large errors equal the experimental errors in K_2 and K_3 . Whether or not the three decimal places are all accurate is another question entirely.

We evaluate the relative contributions to K_2 and K_3 from the rotational resonances versus the vibrational-rotational resonances by separately calculating the refraction from all infrared resonances at frequencies in excess of 1000 cm^{-1} (corresponding to a $10\text{-}\mu\text{m}$ wavelength) and separately for all resonances at frequencies less than 1000 cm^{-1} . The arbitrary boundary of 1000 cm^{-1} , which lies in the $10\text{-}\mu\text{m}$ window, suffices to separate the radio refraction caused by the vibrational-rotational transitions at higher frequencies from the pure rotational transitions at lower frequencies. We find that nearly all of both K_2 and K_3 is caused by rotational resonances. The vibrational-rotational resonances produce only

TABLE 1. Values of K_2 and K_3 for Radio Frequencies

	K_2	K_3
Boudouris	72 ± 10.5	$(3.754 \pm 0.03) \times 10^5$
Birnbaum and Chatterjee	71.4 ± 5.8	$(3.747 \pm 0.029) \times 10^5$
Thayer	64.79 ± 0.08	$(3.776 \pm 0.004) \times 10^5$
Calculated	98 ± 1	$(3.583 \pm 0.003) \times 10^5$

Values of K_2 are in degrees Kelvin per millibar; values of K_3 are in degrees Kelvin squared per millibar.

5.8% of the infrared contribution to K_2 and decrease K_3 by only 0.0009%. Specifically, for the fit to the temperature range 0° – 60°C , the infrared water vapor resonances at $\nu_i > 1000\text{ cm}^{-1}$ give a contribution to K_2 of $1.92 \pm 0.02\text{ K/mbar}$ and a contribution to K_3 equal to $-30.9 \pm 5\text{ K}^2/\text{mbar}$. Notice that the net refraction by these resonances is positive. We have no objection to a negative contribution to K_3 because we do not force a classical interpretation on the refraction by the vibrational-rotational resonances.

We have investigated forcing the rotational resonances to contribute to only one term in (1). For the temperature range 0° – 60°C , we fitted the calculated refraction from all resonances having $\nu_i < 1000\text{ cm}^{-1}$ to the form $K'_2(e/T^p)$ and obtained

$$K'_2 = (3.1748 \pm 0.0083) \times 10^5\text{ K}^p/\text{mbar} \quad (6)$$

$$p = 1.97421 \pm 0.00046 \quad (7)$$

The errors are defined analogous to the errors stated in (4) and (5). This one-term fit gave an rms error of a few thousandths of one percent in the refraction; this is roughly the same error as obtained by fitting with the two terms in (1). We fitted the calculated refraction from all resonances $\nu_i > 1000\text{ cm}^{-1}$ to the form $K'_2(e/T^q)$ and obtained

$$K'_2 = 1.319 \pm 0.038\text{ K}^q/\text{mbar} \quad (8)$$

$$q = 1.0565 \pm 0.0050 \quad (9)$$

This two-parameter, one-term fit gave a 0.02% rms error in the refraction caused by those resonances having $\nu_i > 1000\text{ cm}^{-1}$. In comparison, the two-parameter, two-term fit in (1) gave 0.03% rms error.

DISCUSSION

Table 1 gives the values of K_2 and K_3 resulting from measurements by Boudouris [1963], the review of measurements, including their own, by Birnbaum

and Chatterjee [1952], the values reported by Thayer [1974], who assumed K_2 equals its value at optical frequencies, and the calculations described in the previous section. Birnbaum and Chatterjee [1952] collected the K_2 and K_3 values from several experiments; they averaged the K_2 values weighted inversely proportional to the square of the probable errors and averaged the K_3 values by the same method. The values of K_2 and K_3 given by Smith and Weintraub [1953] are based on those by Birnbaum and Chatterjee [1952]. On the basis of their recent measurement of water vapor refraction, Liebe et al. [1977] give values of K_2 and K_3 that are nearly identical to those of Smith and Weintraub [1953]. The appendix discusses the significantly different values of K_2 and K_3 obtained by Hasegawa and Stokesberry [1975]. The experimental values of K_2 and K_3 seem consistent and well-represented by the upper two entries in Table 1.

Now compare the experimental values in Table 1 (those of Boudouris and of Birnbaum and Chatterjee) with the calculated values. Similar to the calculations by Zhevakin and Naumov [1967a], our calculated refractive index is within a few percent of measured values. At -20°C , the refractivity from our calculated values of K_2 and K_3 is 2.65% less than that obtained using the K_2 and K_3 values of Boudouris. This underestimate decreases with increasing temperature to 0.76% at 260°C . At 20°C our calculated K_2 and K_3 values give 2.4% less refractivity than K_2 and K_3 from Boudouris. For 20°C , Table 2 by Boudouris [1963] summarizes experimental error in refractivity for nine experiments; these errors average to 1.2%. This is one-half the discrepancy between calculated and measured refractivity at 20°C . However, the apportionment between the coefficients K_2 and K_3 differs from the measured results by several standard deviations. For instance, assuming that K_2 has an average experimental standard deviation of 8 (see Table 1), then the experimental value of K_2 is 3.2 standard deviations smaller than its calculated value. Likewise, the experimental value of K_3 is 5.7 standard deviations larger than its calculated value. Thus if one plots $(n-1)/(e/T)$ as a function of T^{-1} , then the calculated curve (a straight line to excellent approximation) is significantly different from the measured curve because the slope K_3 and intercept K_2 of the straight line fits differ by many standard deviations. This is a genuine disagreement between theory and experiment.

One possible explanation for the discrepancy is

that the line parameter compilation is incorrect, in that S_i/v_i^2 is inaccurate for some lines. Considering our previous comments (see the introduction) on *Van Vleck's* [1932] derivation, the rotational resonances most likely to provide a deviation from the $K_3(e/T^2)$ dependence are those having frequencies near $\nu_i \approx K_3 T/hc \approx 190 \text{ cm}^{-1}$ (i.e., 5.7 THz). Since the term $K_2(e/T)$ is much smaller than the $K_3(e/T^2)$ term, even a small deviation from a $1/T^2$ dependence by the rotational contribution will cause a large contribution to K_2 .

A second possible explanation of the discrepancy is that water clusters contribute to the measured refraction, whereas we have only calculated the monomer contribution. In principle, it is possible to calculate the refraction contributed by a model absorption spectrum of the water dimer (the dimer absorption spectrum is given by *Bohlander* [1979]). However, it is not possible to calculate the refraction contributed by the broadband empirical excess absorption, for two reasons. First, the Kramers-Kronig relation is sensitive to the way in which this absorption decreases at high frequencies. Second, the excess absorption is defined as the measured absorption less the monomer absorption predicted with a particular line shape. To the extent that the tails of this particular monomer line shape are inaccurate, the excess absorption cannot be entirely attributed to water clusters; but one must accurately remove monomer absorption from the excess absorption because the refraction contribution of the monomer is fully accounted for in our calculations.

The discrepancy is not caused by our calculation using a 1-m radio wavelength (300 MHz) as opposed to the higher radio frequencies typically used in experiments (see Table 2 by *Boudouris* [1963]). The dispersion in $(n-1) \times 10^6$ and A_T is shown graphically by *Hill and Clifford* [1981], where $A_T \approx -K_3(e/T^2)$; thus the dispersion in A_T is a measure of that in K_3 . The dispersion in K_2 is easily calculated from that in $(n-1) \times 10^6$ and K_3 . From zero frequency to 60 GHz the graphs by *Hill and Clifford* [1981] imply that K_3 increases by $0.0096 \times 10^5 \text{ K}^2/\text{mbar}$ and K_2 decreases by 1.6 K/mbar. These changes in K_2 and K_3 are considerably smaller than their experimental uncertainties.

Now consider Thayer's values of K_2 and K_3 in Table 1. Those values are based on his hypothesis that K_2 should be the same at radio wavelengths as at visible wavelengths, namely $K_2 = 64.79 \text{ K/mbar}$. We have seen, however, that the infrared resonances

of water vapor are theoretically expected to give a contribution to K_2 ; and the calculated values of K_2 show that the infrared resonances of water vapor can give a significant contribution to K_2 (namely a calculated contribution of 33.3 K/mbar in (4)). Thus Thayer's hypothesis is unfounded. We suggest that one should use the entirely empirical coefficients (e.g., those of *Boudouris* [1963]). Of course, the term $K_2(e/T)$ is only needed for precise work. For most practical applications, this term may have its approximate value introduced into the $K_3(e/T^2)$ term to yield the simpler radio refractive index equations discussed by *Smith and Weintraub* [1953] or *Bean and Dutton* [1966].

We conclude, quite opposite to the assumption by *Zhevakin and Naumov* [1967a], that the infrared contribution to K_2 is dominated by the rotational resonances, not by the vibrational-rotational resonances. We noted in the introduction that the rotational resonances of water vapor need not provide a temperature dependence proportional to $1/T^2$. In fact, *Zhevakin and Naumov* [1967a] calculated the temperature dependence of radio refractive index caused by the rotational resonances; they obtained $1/T^{1.97}$. If one fits $1/T^{1.97}$ to the form $(K_2/T) + (K_3/T^2)$, one obtains a value of K_2 that is roughly the magnitude of the K_2 contribution that we found from the rotational resonances. Thus the contribution of the rotational resonances to K_2 is very sensitive to the deviation from $1/T^2$ of their refractive temperature dependence.

One could force the rotational transitions to contribute to only one term in the radio refractive index equation by fitting their radio refraction to $K'_3(e/T^p)$. Then p would be close to, but not equal to, 2 (just as *Zhevakin and Naumov* [1967a] found $p \sim 1.97$). Our fit to this functional form gives the K'_3 and p values in (6) and (7). One would then likewise require that the vibrational-rotational transitions contribute to only one term by fitting their radio refraction contribution to $K'_2(e/T^q)$. Then q would be nearly, but not precisely, unity. Our fit gives the K'_2 and q values in (8) and (9). We must view this type of fit to be in no fundamental sense superior to that in (1), and this fit obscures comparison of the calculated and measured refractive indexes. Since the rotational transitions do not provide K_3 exclusively and the vibrational-rotational transitions do not contribute to K_2 exclusively, we must view (1) as a low-order polynomial fit in inverse powers of T to a general function of temperature.

In fact, *Van Vleck* [1927] shows that the quantum mechanical correction to (1) has the form $K_4(e/T^3) + K_5(e/T^4) + \dots$. By including the term $K_4(e/T^3)$ in (1) and using *Van Vleck's* expression for K_4 , *Birnbaum and Chatterjee* [1952] (in their appendix) show that the value of K_2 is reduced from its value deduced using (1) without the correction term. They show that this reduced value of K_2 is less than the extrapolation to zero frequency of the optical-frequency value of K_2 ; this is impossible since the (positive) infrared contribution to K_2 must be added to its extrapolated optical value to obtain the radio frequency value of K_2 . Since *Van Vleck's* derivation assumes that there are no strong resonances having energy changes of the order of the thermal energy $K_B T$ and since we have seen in the introduction that the water molecule violates this assumption, it follows that *Van Vleck's* value for K_4 should not be applied to water vapor and the discrepancy noted by *Birnbaum and Chatterjee* [1952] is removed.

Summing the refraction by absorption resonances can shed light on the radio refraction of dry air as well as water vapor. *Liebe* [1981] calculates refraction by oxygen resonances having resonance frequency less than 1000 GHz. These magnetic dipole transitions of molecular oxygen are expected to be the main contributors to the magnetic susceptibility of air at radio frequencies. *Liebe's* calculated refraction caused by these oxygen resonances accounts for three fourths of the observed magnetic susceptibility of air; reasons for the remaining difference in calculated versus observed susceptibility are unknown.

APPENDIX

Hasegawa and Stokesberry [1975] obtained values of K_2 and K_3 that are significantly different from those of *Boudouris* [1963] and *Birnbaum and Chatterjee* [1952]. *Hasegawa and Stokesberry* [1975] used the *Birnbaum-Chatterjee* method of averaging K_2 and K_3 from several different experiments. Unlike *Birnbaum and Chatterjee*, *Hasegawa and Stokesberry* included experiments that covered only very limited ranges in temperature, namely the two experiments by *Essen and Froome* [1951] and *Essen* [1953]. Because of their limited temperature range, *Essen and Froome* [1951] assumed K_2 equal to its value at optical frequencies (an inaccurate assumption) and fitted only K_3 to their data. Because of their small probable error, these two experiments

overwhelmingly dominated the average K_2 and K_3 determined by *Hasegawa and Stokesberry* [1975]. Because of the very limited temperature range of these two experiments and the inappropriate assumption regarding K_2 , we feel that the average values of K_2 and K_3 of *Hasegawa and Stokesberry* are not accurate. It seems preferable, but perhaps impossible, to fit (1) to a compilation of available experimental values of refractivity, rather than to average the coefficients of fits made to each individual data set.

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